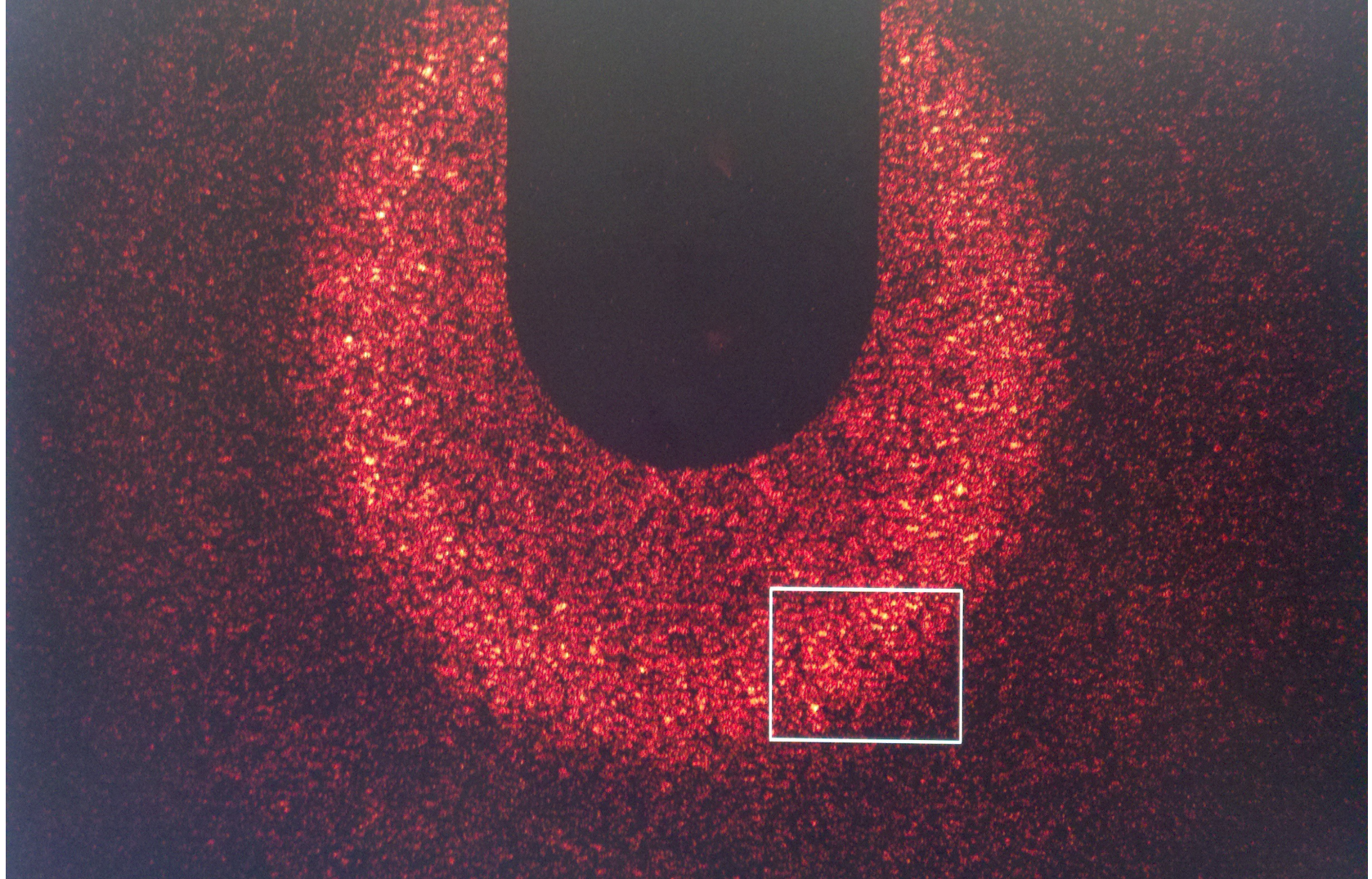


SVS at LCLS

Speckle Visibility/Variance Spectroscopy at the Linac Coherent Light Source (XCS Beamline)

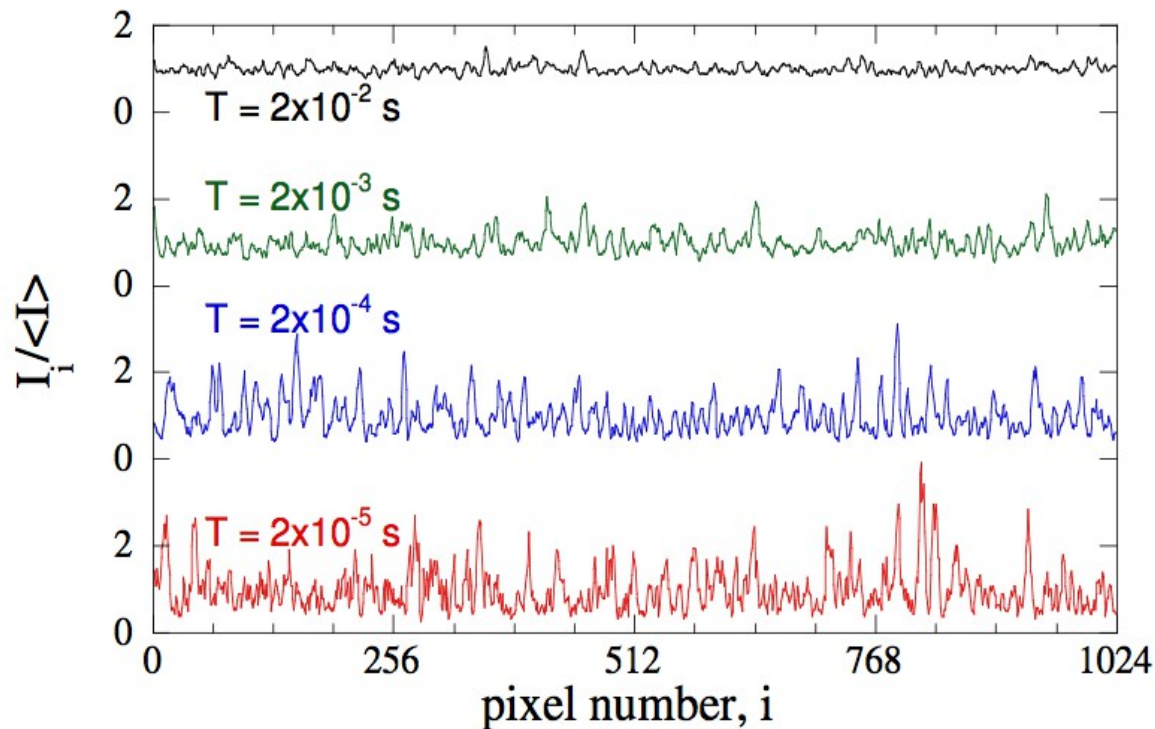


Why SVS?

- Due to technological limitations, detector readout times may exceed timescale of sample dynamics.
- For example, with FEL split-and-delay schemes, no current detectors will be able to separately read out pulses separated by picoseconds.
- Instead of reading out multiple exposures separately, SVS analyzes statistics of speckle patterns as a function of exposure time.
- At LCLS, there were problems running the CCD in “kinetics mode,” and the long minimum readout time of about 5 seconds made it seem an attractive option.

Basic Principle of SVS

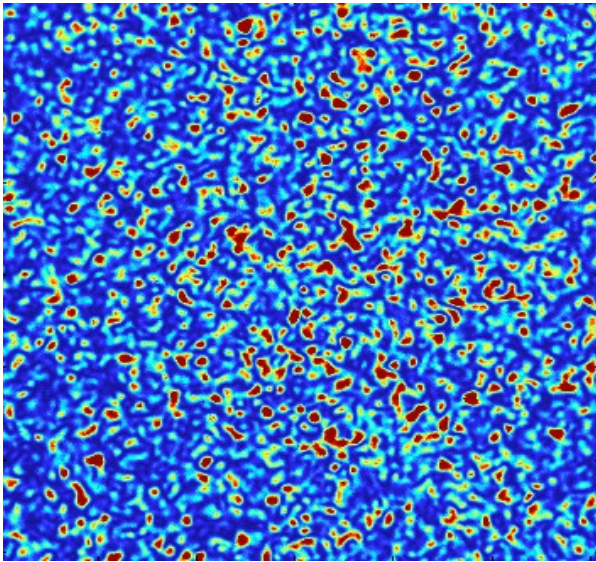
- When dynamics are present, longer exposures result in reduced contrast (variance of intensity).



From:
Bandyopadhyay et al.(2005)

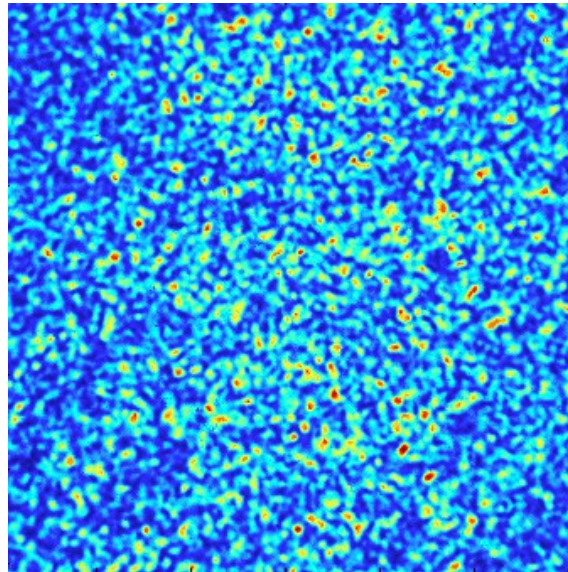
- By looking at the variance of the image intensity vs. exposure time, the autocorrelation function can be determined.

A contrived example: scotch tape in the laser lab

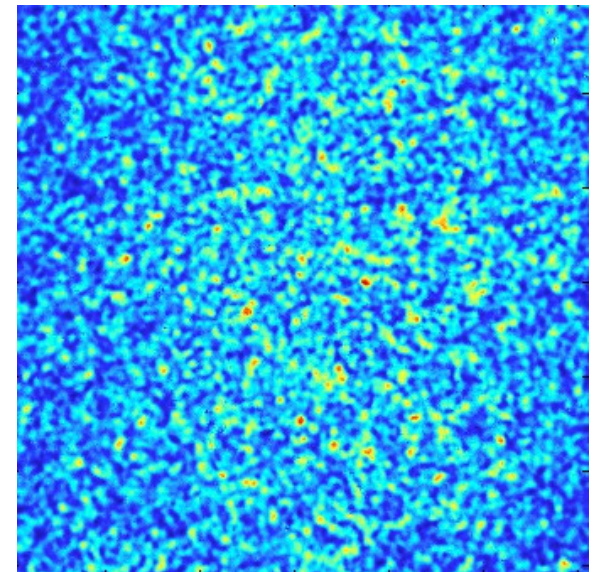


First frame

Each frame is a snapshot of a static sample, but a different region is illuminated for each frame. Therefore, each of the images are uncorrelated.



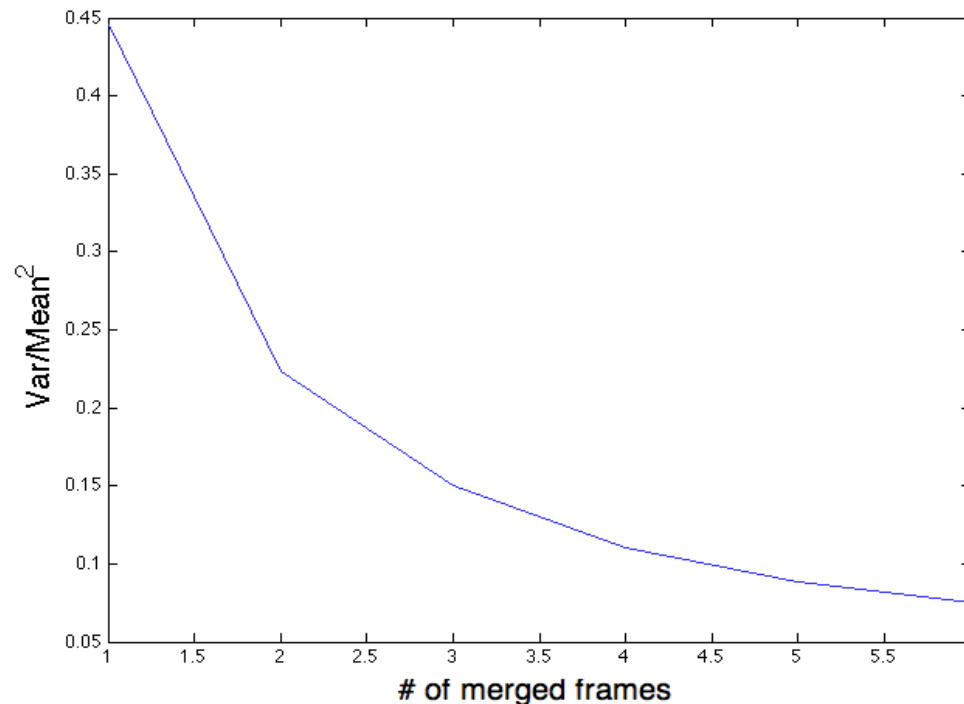
First 3 frames



First 6 frames

How does this become quantitative?

In SVS, the quantity of interest is $\text{Var}(I) / \langle I \rangle^2$, the ratio of image variance to the mean squared. We want to know how this depends on exposure time.



← This plot is for the scotch tape speckle patterns.

Bandyopadhyay et al.(2005) show that this quantity is related to the autocorrelation function as follows...

The Fundamental Equation of SVS

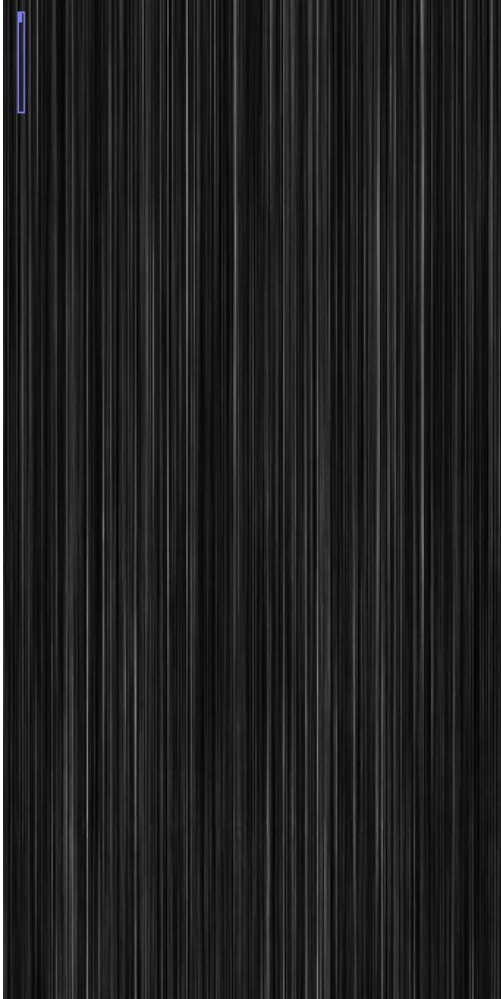
$$V_2(T) = \frac{1}{\beta} \left[\frac{\langle I^2 \rangle_T}{\langle I \rangle^2} - 1 \right] = \int_0^T 2(1 - t/T) [g_1(t)]^2 dt / T$$

Speckle Visibility = A weighted average of the (electric field) autocorrelation function

- One specifies a functional form for g_1 (e.g. exponential) and fits to data to estimate parameters (e.g. tau).
- This can be done for higher moments as well (v_3 , etc.).
- Note that V_2 will in general fall off less quickly than g_1 . This is another advantage of SVS, as it makes faster timescales accessible with an otherwise equivalent experimental setup.

Collapsing Foam in Laser Lab

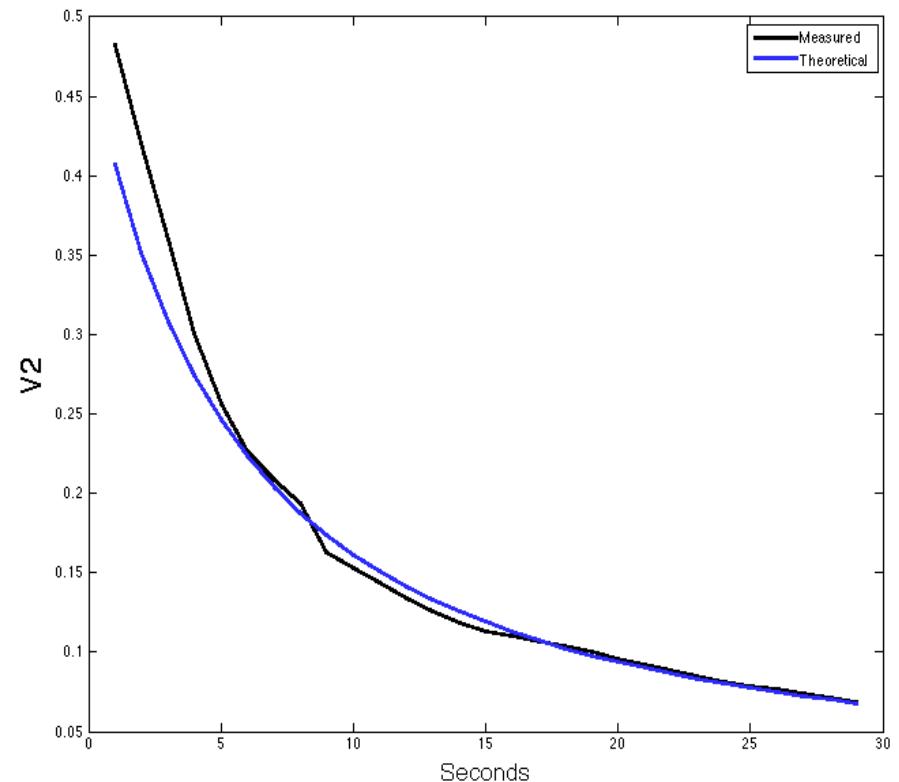
Here is a line scan image of speckle from shaving cream. Each row of pixels corresponds to one exposure of 900 microseconds. The entire image is 32,768 lines vertically = about 30 seconds total, but only about 1/10 of image is shown here.



According to the literature,
for multiple scattering from
shaving cream in
transmission,

$$g_1(t) = \sqrt{6\Gamma t} / \sinh \sqrt{6\Gamma t}$$

Numerically integrating
this according to the
SVS equation, choosing
an appropriate gamma,
and plotting alongside
the computed values of
V2 gives →

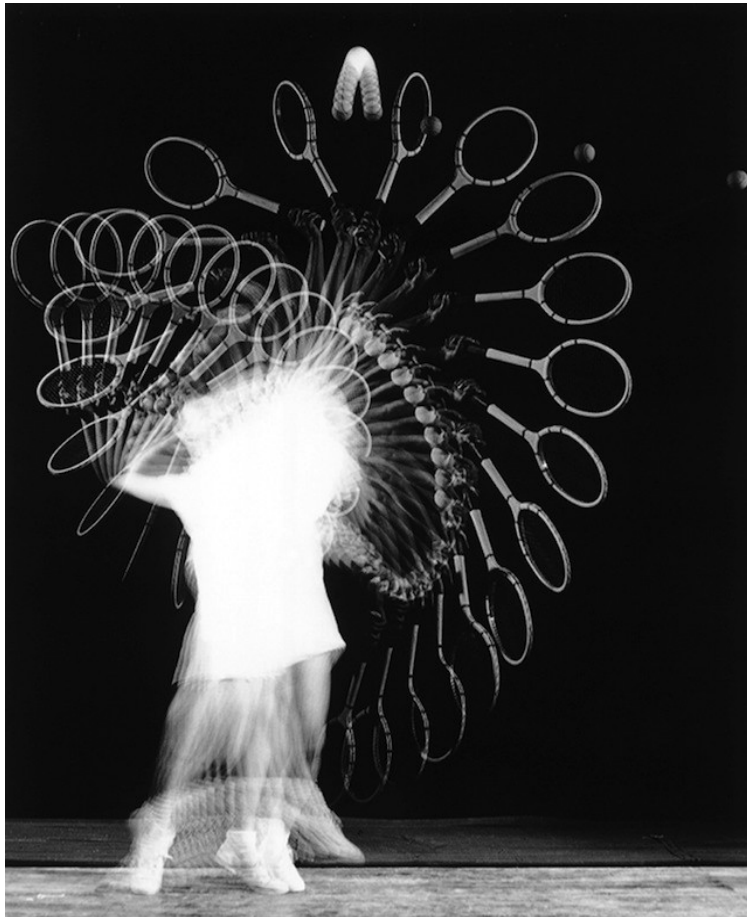


Now, on to LCLS...

A free-electron laser (FEL) is essentially an x-ray strobe light. (So the SVS integral needs to become a discrete sum.) There are 3 times of importance:

- 1) Length of x-ray pulses (about 100 femtoseconds).
- 2) Time between pulses (8.3 milliseconds)
- 3) Time between images (several seconds)

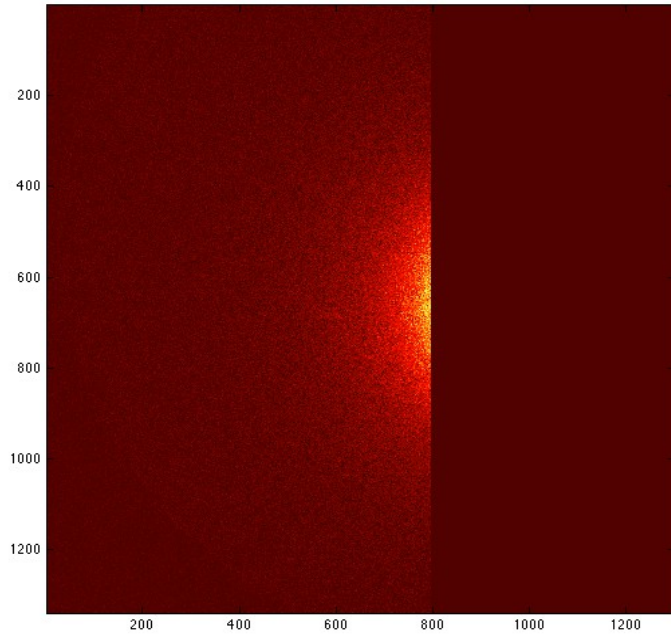
LCLS



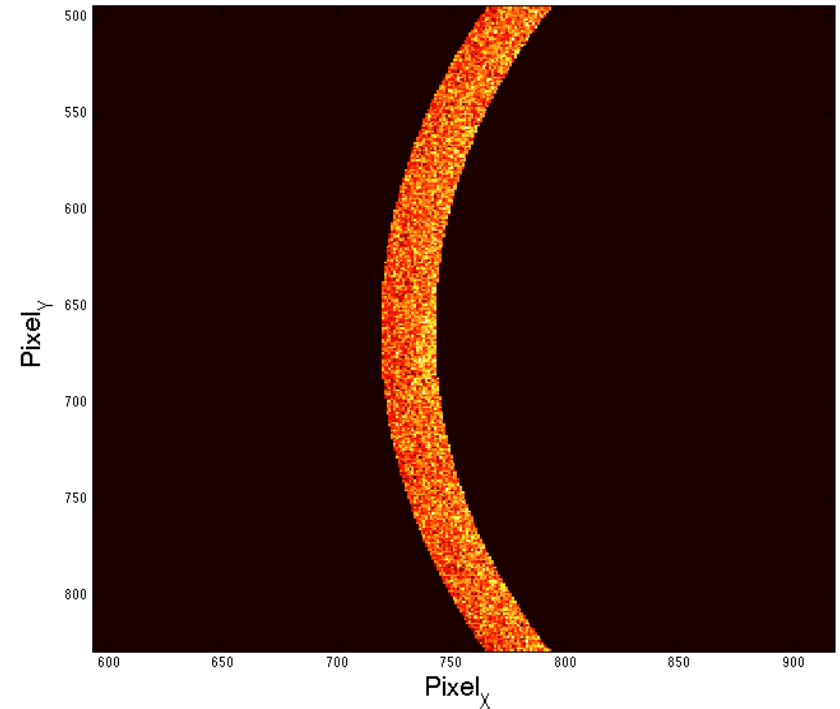
“C-W”
Source

(Images from Google)

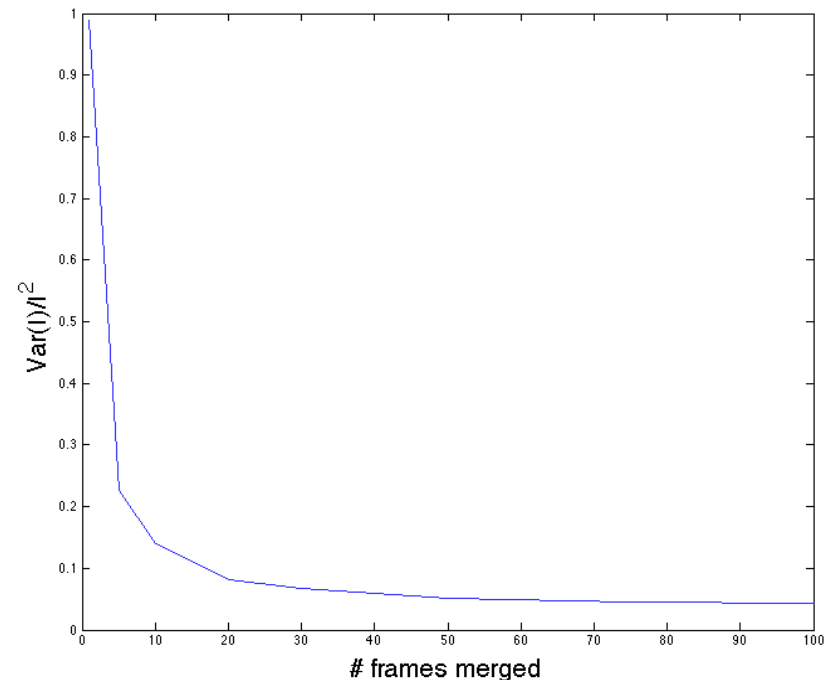
Analyzing LCLS data



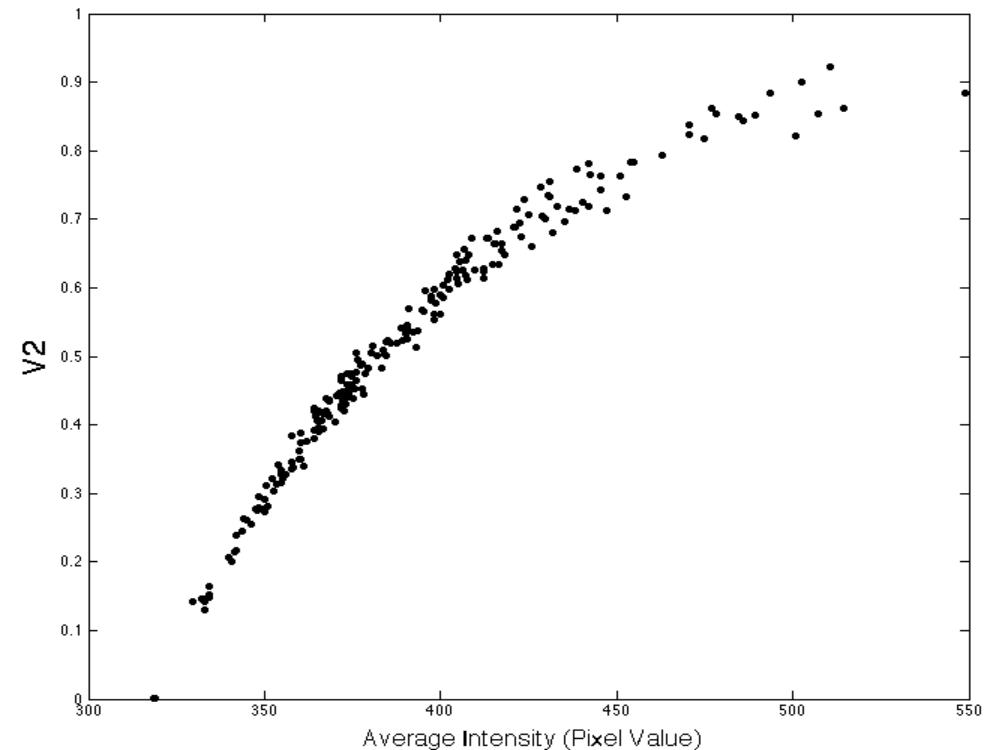
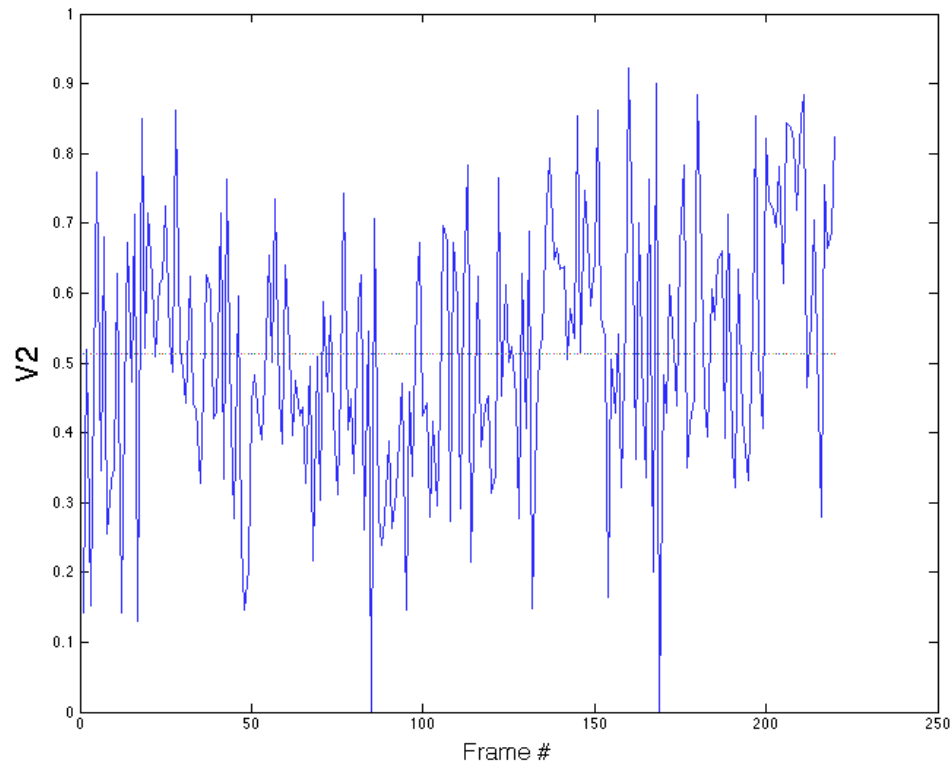
→ Choose an annulus (q region).



→ Plot V_2 versus number of merged frames. In this particular case, each frame consisted of 10 x-ray “pulses” (chosen by researchers), each separated by 8.3 ms (120 Hz). The pulses are about 100 fs long. The individual frames were separated by about 5 sec (limited by detector readout speed).

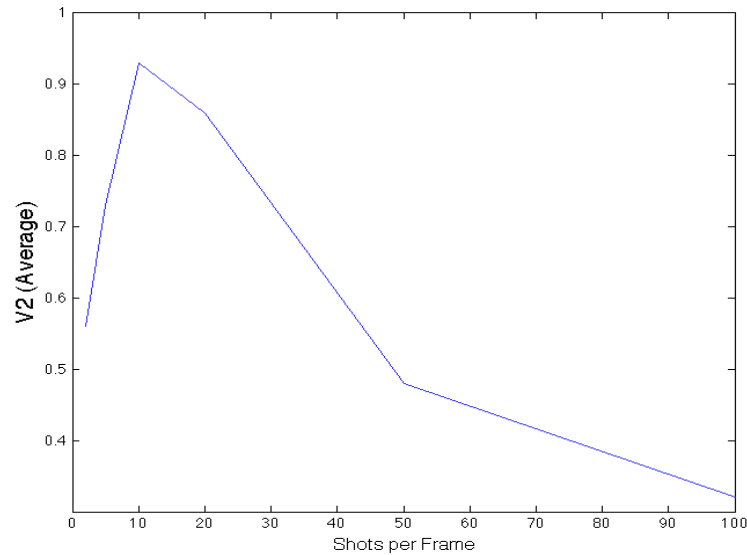


At LCLS, the intensity of each “shot” varies considerably from the average. When there are low numbers of shots per frame, this creates a large variability in V2 between frames as well.

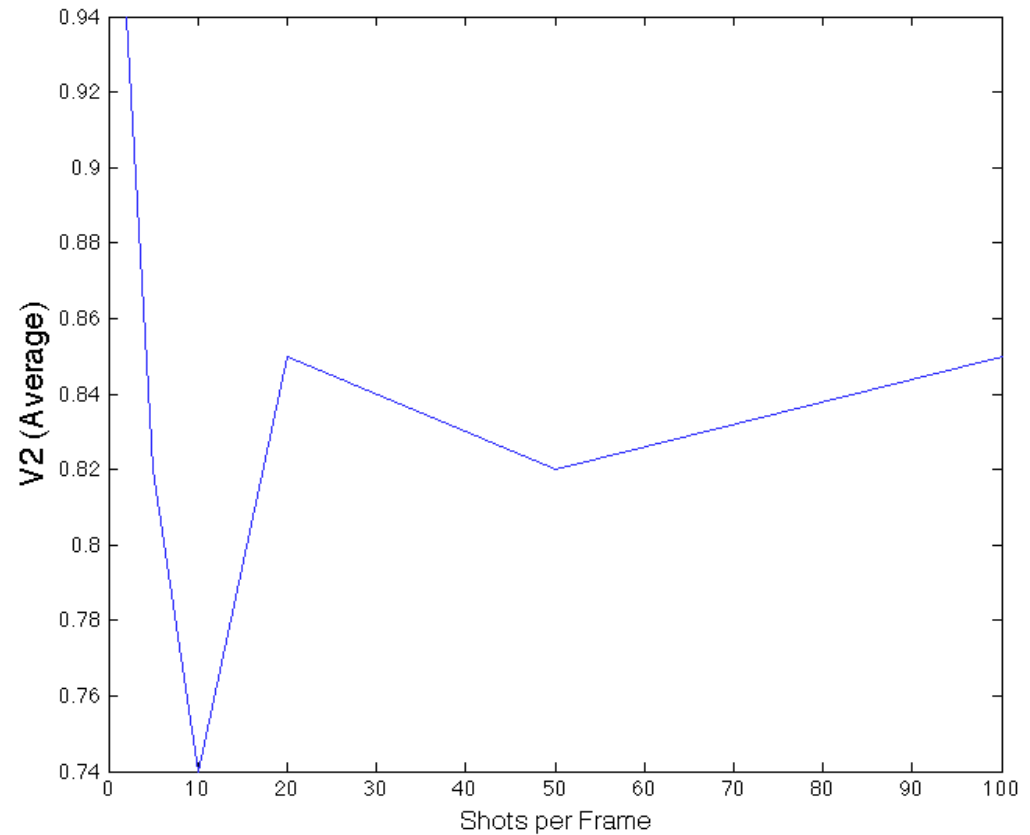
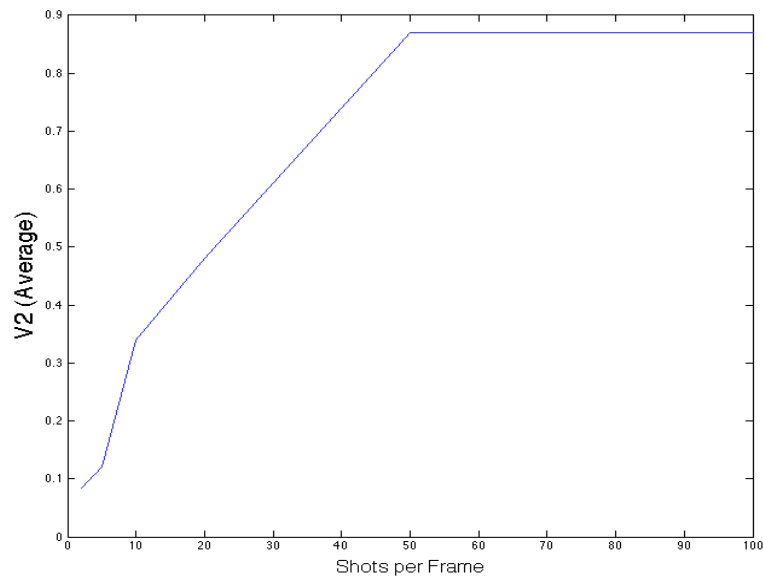


The above sample was Au nanoparticles(NPs) in polystyrene(PS) 42k @ 170 C. Two shots per frame. The first graph shows V2 computed for each frame, and the second shows V2 vs. the average intensity of that frame.

How does V2 depend on shots per frame?

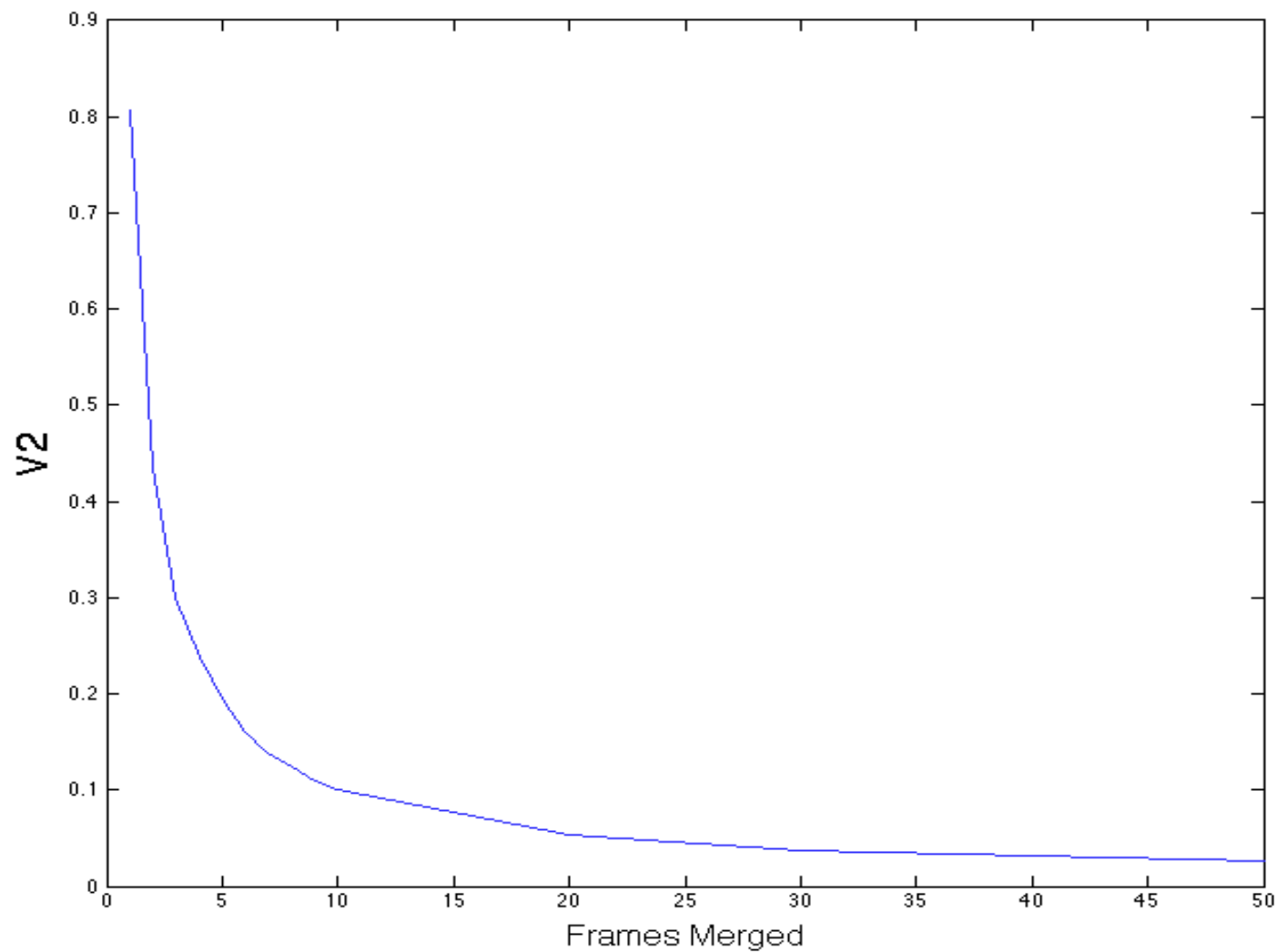


Au-NPs in PS 42K @ 170C.
 $Q=0.05 \text{ \AA}^{-1}$ above, 0.1 below.



Dry silica at room temperature.
 $Q=0.05 \text{ \AA}^{-1}$. This was supposed to be a static reference sample. *But if we look at V2 vs. frames merged...*

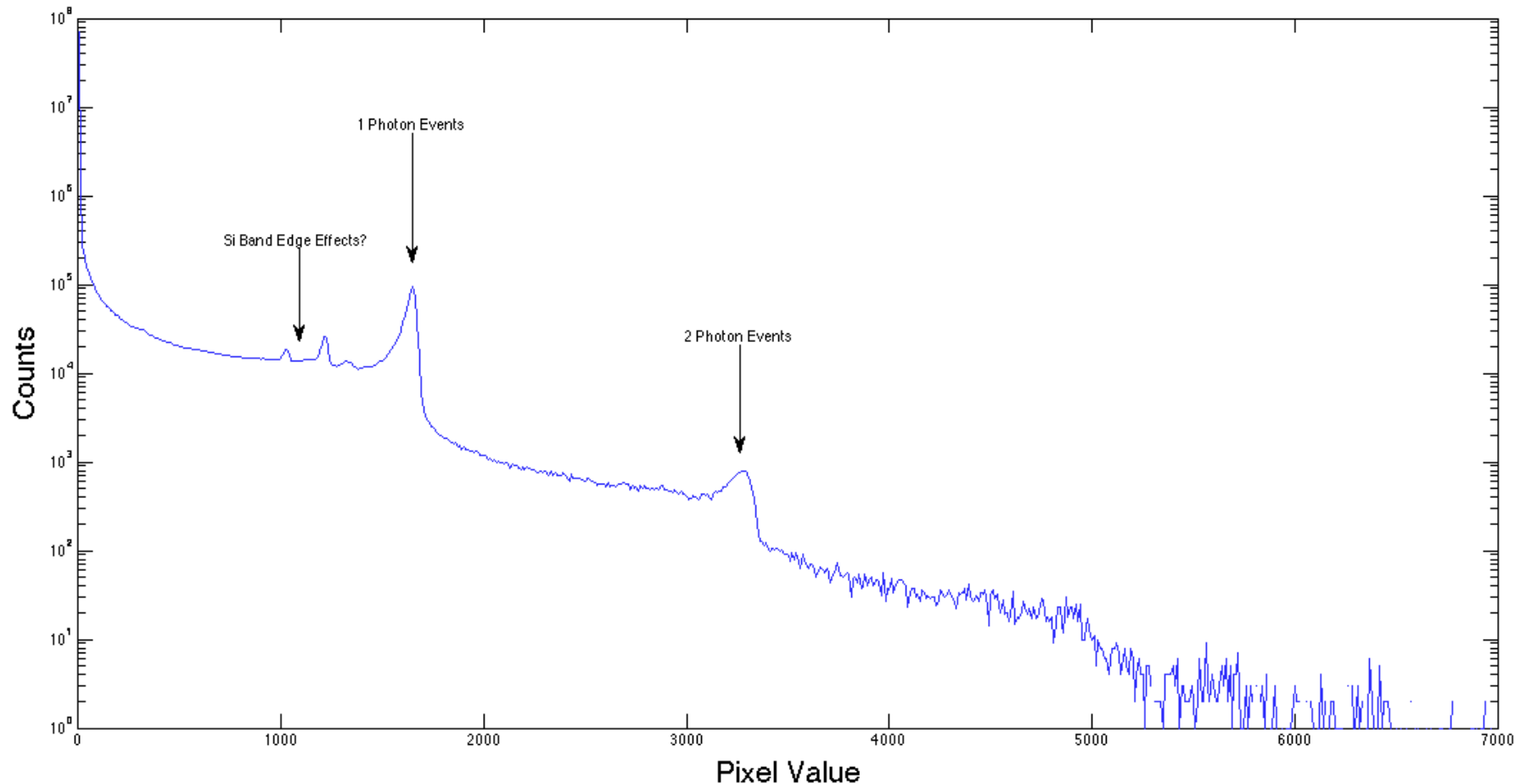
...V2 falls off much too fast



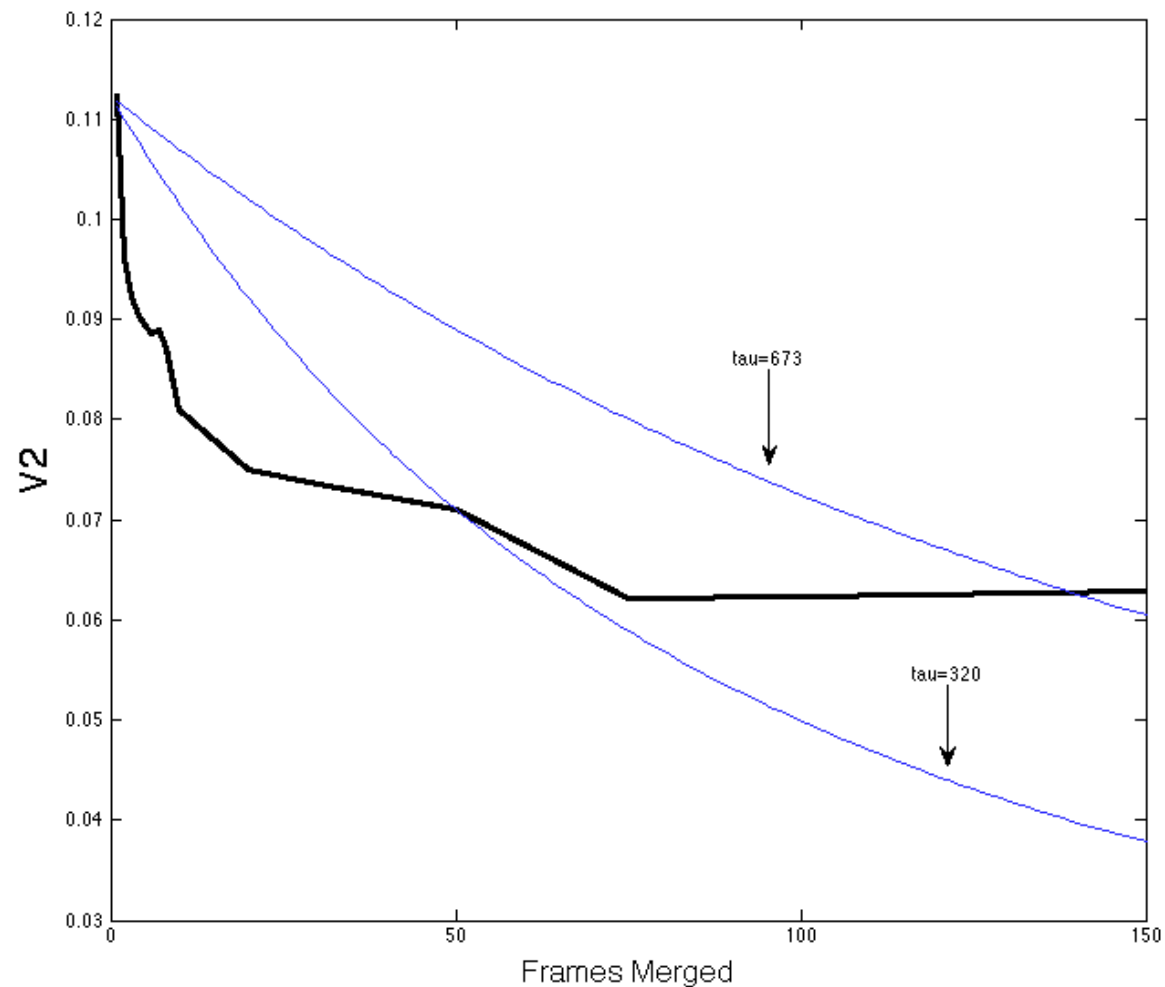
(Silica with 20 shots per frame.)

Histogram of Pixel Values

With a fully coherent source, and sufficient photon counts, the probability distribution of speckle intensity should be exponential (which gives the theoretical upper bound of 1 on V_2). However, the distribution of actual pixel values is not exponential in the low photon limit.



And for samples with dynamics, V2 doesn't have the proper shape...

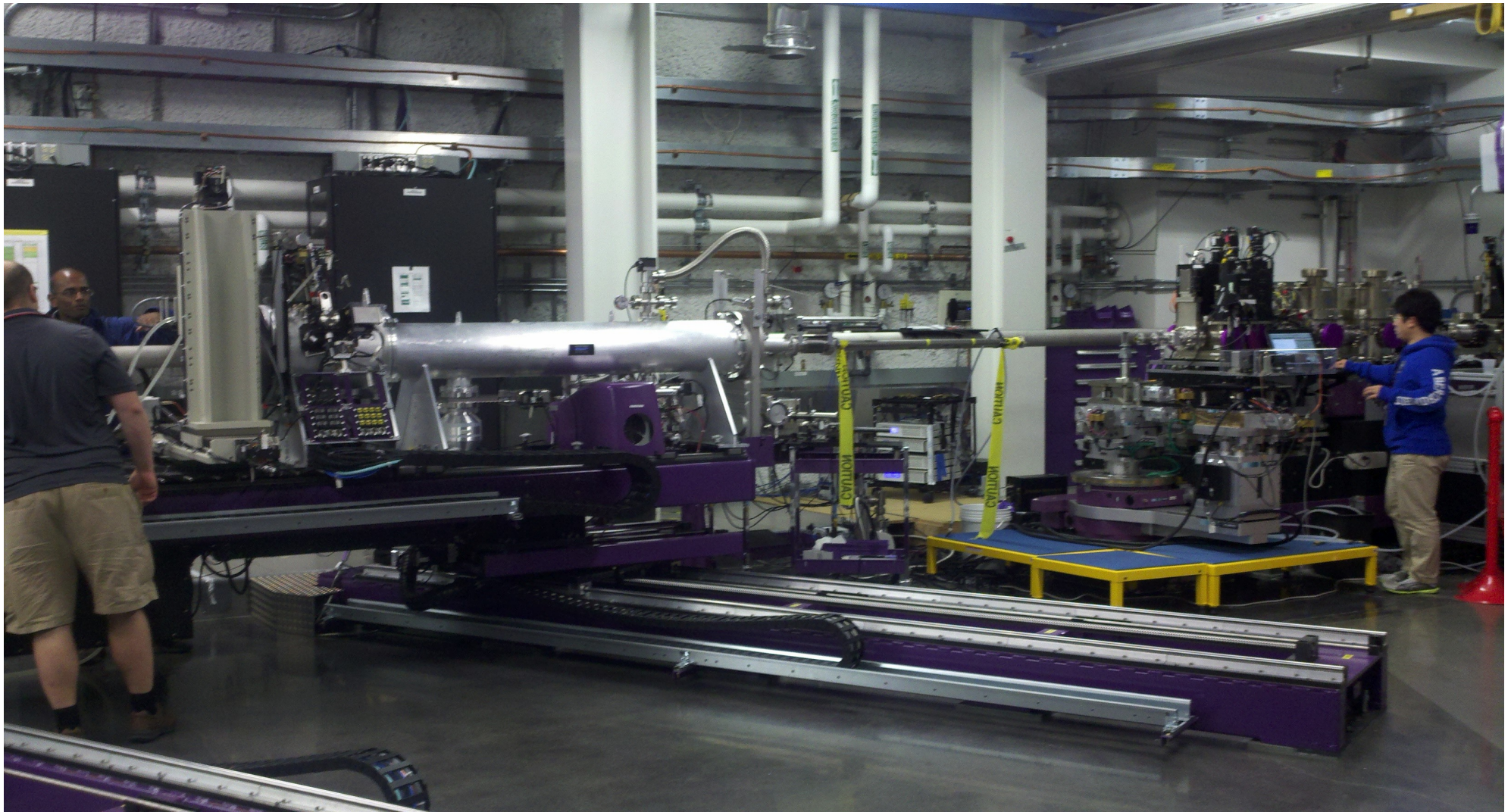


The above sample was Au nanorods in PS1000k. Traditional autocorrelation fitting produced a τ of 673 sec. Beam jitter from *frame to frame* would explain the rapid initial fall-off. But from previous graphs, jitter is clearly not an issue from *pulse to pulse*.

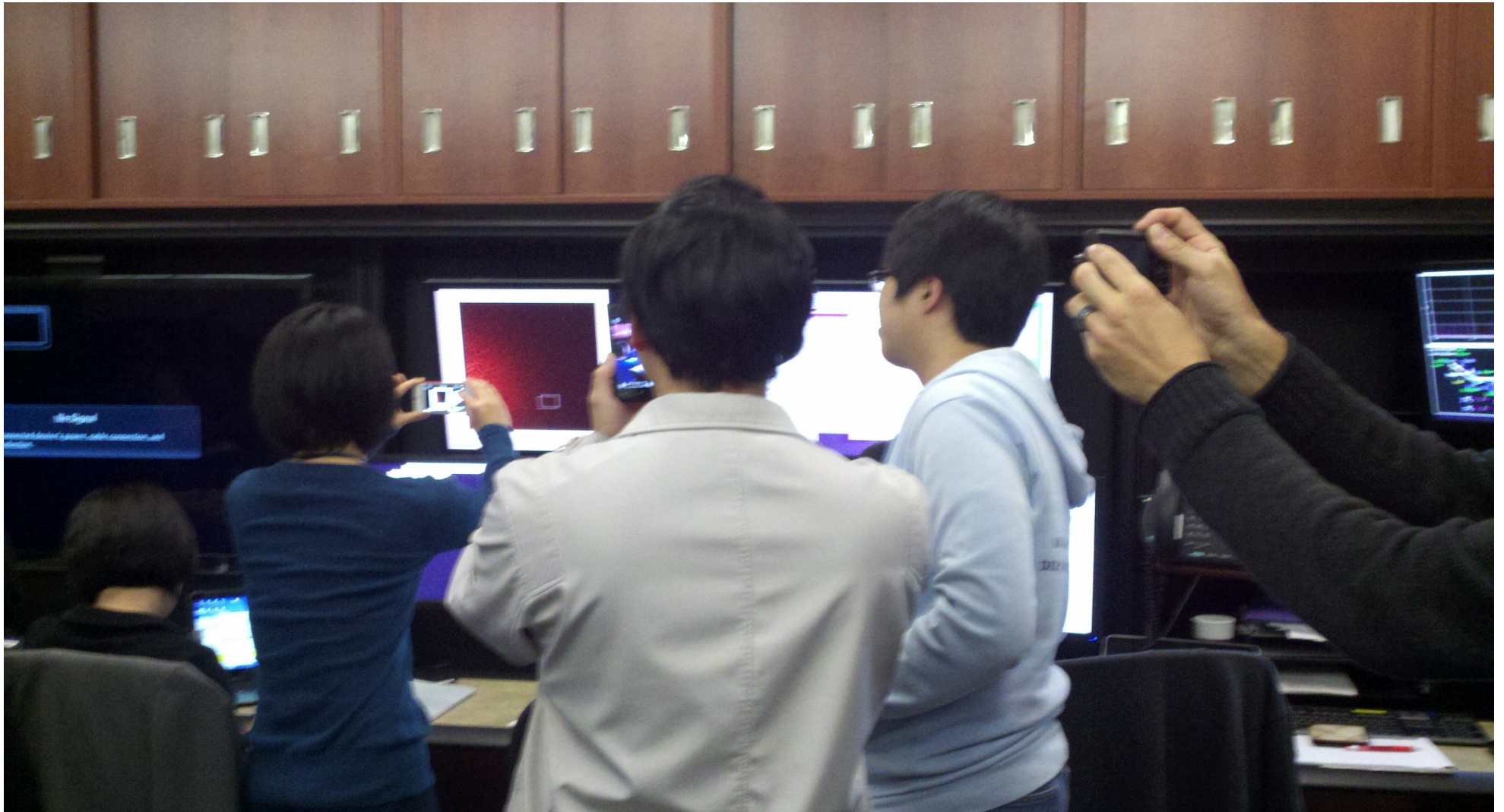
And now for some pictures...



The Far Experimental Hall (FEH), home of three beamlines: XCS (x-ray correlation spectroscopy), MEC (matter in extreme conditions), XCI (x-ray coherent imaging).



Inside the Hutch. Big enough for several rabbits.



Nested images of the first speckle pattern.



Oleg sneaking a photo of the klystron gallery, without a dosimeter!



The huge pep ring detector across from the FEH. The upper third of this picture is above ground, and is the size of a big airplane hanger.



On the last day, it was finally dry out, and the boss and I got to SLACK off a bit...